

Homometry in the light of coherent beams

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Two systems are homometric if they are indistinguishable by diffraction. We first make a distinction between Bragg and diffuse scattering homometry, and show that in the last case, coherent diffraction can allow the diffraction diagrams to be differentiated. The study of the Rudin-Shapiro sequence, homometric to random sequences, allows one to manipulate independently two-point and four-point correlation functions, and to show their effect on the statistics of speckle patterns. Consequences for the study of real materials are discussed.

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INTRODUCTION

The possibility to shape coherent X-ray beams from synchrotron light sources [1] and to get naturally coherent beams from X-ray free electron lasers (XFEL) [2], has revolutionized the way X-ray diffraction experiments are performed and analyzed. One of the most fascinating property of coherent diffraction is the possibility to measure speckle patterns [3], which are much more informative than the diffuse scattering obtained by classical diffraction. Together with the development of novel sources, phase retrieval algorithms have also emerged, allowing the reconstruction of the diffracting objects under certain conditions [4, 5]. However, the reconstruction of a structure is not always possible nor necessary to study the physics of materials. For example, measuring correlation lengths close to phase transitions [6] or slow dynamics with X-ray photon correlation spectroscopy [3, 7] does not require the full reconstruction of the system under study.

The purpose of this letter is to show that statistical analysis of speckle patterns can yield information on orders hidden to conventional X-ray analysis. In this respect, we are in line with recent works showing that four-point intensity cross-correlation of speckle patterns can uncover "hidden symmetries" present in colloidal glasses [8, 9] or magnetic systems [10].

Our approach uses the concept of homometry, *i.e.* the property of different systems to exhibit same diffraction patterns. Separating out the scattered intensity expression into three terms allows one to show that homometry can occur at different levels. We then put the emphasis on diffuse scattering homometry, that we discuss with the help of the well-known Rudin-Shapiro sequence [11, 12].

COHERENT DIFFRACTION

Let us first give a general expression of the intensity scattered at scattering vector q , by a 1D periodic N -site lattice decorated by two atoms A and B , of scattering factor f_A and f_B , in proportion x and $1 - x$ re-

spectively. Generalization to 2D, 3D, multi-atomic basis, displacement disorder, or disorder of the second kind [13] is straightforward. Following ref. [13], the diffracted intensity is given by the formulae:

$$I(q) = \sum_{n,n'} f_n f_{n'} e^{iq(n'-n)} = \sum_m \sum_n f_n f_{n+m} e^{iqm}. \quad (1)$$

The *ensemble* average of the cross-product $\langle f_0 f_m \rangle$ is then introduced:

$$\frac{1}{N_m} \sum_n f_n f_{n+m} = \langle f_0 f_m \rangle + \Delta_m, \quad (2)$$

where N_m is the m -dependant number of terms of the sum \sum_n . The Δ_m term, usually neglected in textbooks, is due to finite-size fluctuations of the spatial average with respect to the ensemble one.

Further introduction of $\Delta f_m = f_m - \langle f \rangle$ allows one to get the three components of kinematic diffraction:

$$I_B(q) = \langle f \rangle^2 \sum_m N_m e^{iqm} \quad (3a)$$

$$I_{DD}(q) = \sum_m N_m \langle \Delta f_0 \Delta f_m \rangle e^{iqm} \quad (3b)$$

$$I_S(q) = \sum_m N_m \Delta_m e^{iqm} \quad (3c)$$

The first term gives the intensity of the Bragg reflections, and the fringes due to finite size effects. For a crystal of N cells of structure factor $F(q)$, it can be written as:

$$I_B(q) = |\langle F(q) \rangle|^2 \frac{\sin^2 q(N+1)/2}{\sin^2 q/2} \quad (4)$$

In practice, the fringes given by the sine functions are only visible with a coherent beam illumination (for conditions of observation and examples see [3]).

The second term is the diffuse scattering intensity, which only depends on pair correlation function (CF). For *random* disorder, it reduces to the well-known Laue formula:

$$I(q) = Nx(1-x)(f_A - f_B)^2, \quad (5)$$

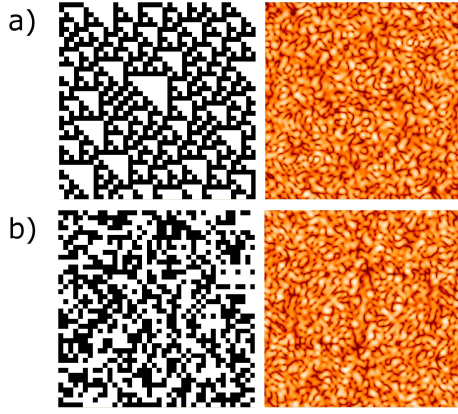


FIG. 1. (left) (40×40) lattices of $+1$ and -1 in equal proportion and (right) diffraction patterns in the first Brillouin zone (log scale) for a) a triplet SRO lattice [14] b) a random lattice. Reciprocal nodes are in the corners.

where *random* refers to the vanishing of the pair CF (*i.e.* $\langle \Delta f_0 \Delta f_m \rangle = \langle \Delta f_0 \rangle \langle \Delta f_m \rangle = 0$ for $m \neq 0$.)

The third term gives rise to speckles. Like fringes, speckles only exist if the incident beam is coherent enough, *and* if the system does not explore too many configurations during acquisition time T (non-ergodicity condition $\langle \Delta_m \rangle_T \neq 0$). Interestingly enough, working out of coherent conditions has the effect of averaging out Δ_m , which yields *ensemble* averaged quantities.

HOMOMETRY

Homometry - etymologically *same distance* - is a word coined by A. Patterson [15, 16], to describe the property of non-congruent sets of points to possess the same pair distances (or the same difference sets) [17]. Homometric sets have thus the same diffraction pattern, as Eq. (1) demonstrates. A simple example of homometry is given by the two sets $S = \{0, 1, 4, 10, 12, 17\}$ and $S' = \{0, 1, 8, 11, 13, 17\}$ [17]. Indeed, the structure factors $F(q)$ of the two sets have the same magnitude for *all* q -vectors, but not the same phase. Hence, the loss of the phase makes these sets *indistinguishable* by X-ray diffraction.

However, because solid state physics deals with materials, the above definition turns out to be too restrictive. Eqs (3) allows to distinguish between Bragg (B) homometry, diffuse scattering (D) homometry and coherent diffraction (C) homometry.

BRAGG HOMOMETRY

B-homometry describes crystals with different basis but same Bragg intensities [15, 16]. To illustrate that, let us consider the examples of 1D homometric crystals pre-

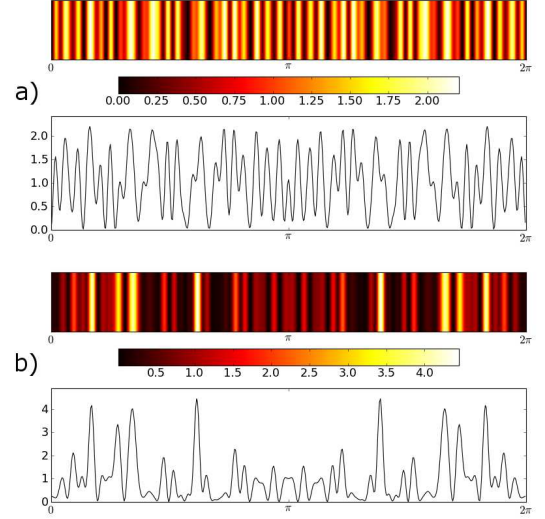


FIG. 2. Diffraction patterns and associated intensity variations from $((N, M) = (64, 512))$ a) RS and b) BS sequences. The vertical broadening of the patterns are for visual convenience.

sented in [16], of unit cells size equal to 8 and atomic positions given by $H = \{0, 3, 4, 5\}$ and $H' = \{0, 4, 5, 7\}$. Structure factors, readily calculated as:

$$F_H(q) = 1 + 2 \cos q + \cos 4q \quad (6)$$

$$F_{H'}(q) = 2(\cos 2q + \cos 4q), \quad (7)$$

have the same amplitude squared *at* the Bragg positions $q = 2\pi h/8$ but not *out of* Bragg positions. Eq (4) shows that the fringes intensity, revealed by coherent diffraction, gives out-of-Bragg values of $|F(q)|^2$ which, at least in theory, allows to distinguish H and H' , and solves the B-homometry issue. This is well-known and corresponds to the oversampling requirement of the phase retrieval algorithms [18, 19]. It is clear however that if the atomic basis is homometric itself, like *e.g.* in crystals with S or S' basis, the problem cannot be solved, coherence or not.

DIFFUSE SCATTERING HOMOMETRY

Surprising examples of D-homometry were designed by Welberry *et al.* [14, 20]. They consist in substitutionally disordered lattices with triplet (or quadruplet) short range ordered (SRO) CF [21], but zero two-point correlations (Fig. 2a). Diffraction diagram of these lattices present the same Bragg and diffuse scattering intensity [14]. But, as shown in Fig. 2, their speckle patterns are different, which breaks the D-homometry.

A more tractable (though subtle) example of D-homometry is provided by the geometrically ordered (GO) [22] Rudin-Shapiro (RS) sequence [23], whose

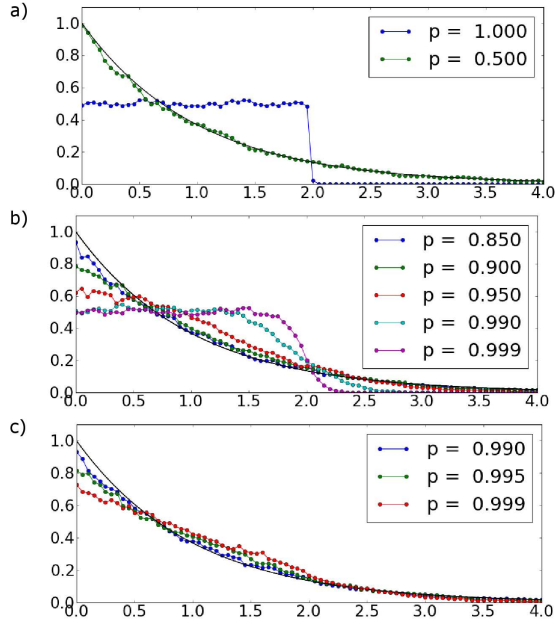


FIG. 3. Probability densities $P(I)$ of diffraction patterns of $(N, M) = (4096, 4096 \times 128)$: a) BS (green) and RS sequence (blue), b) sequences obtained by Bernoullization c) g_4 -SRO sequences for $\xi \sim N/37$ ($p = 0.99$), $N/13$ ($p = 0.95$) and $N/3$ ($p = 0.999$). The line indicates the random negative exponential law.

generic term σ_n can be written [12]:

$$\sigma_{4n+l} = \begin{cases} \sigma_n & \text{for } l = 0, 1 \\ (-1)^{n+l} \sigma_n & \text{for } l = 2, 3 \end{cases} \quad \text{with } \sigma_0 = 1. \quad (8)$$

This sequence has become famous [11, 22–24] because, thought GO, it is D-homometric to randomly distributed sequences (sometimes called Bernoulli sequences (BS) [24]), with the same $4Nx(1-x)$ diffuse scattering intensity (Eq. 5). In other words, its two-point CF $g_2(n) = \overline{\sigma_0 \sigma_n}$ is zero for $n \neq 0$ (spatial average).

Let us now consider the coherent diffraction of RS and BS sequences. In what follows, we present Fast Fourier Transforms (FFT) computations of sequences of length N , zero-padded up to a value $M \gg N$ to clearly see the speckles. For the RS sequence, because $x \neq 0.5$ and depends on its length N , we have always substracted the average value $2x - 1$ to all the terms in order to get rid of the Bragg intensities. BS of 1 and -1 in equal proportion were computed with the python pseudo-random number generator. The squared value of the FFT $I(q)$ are normalized by $4Nx(1-x)$ in order to get $\overline{I(q)} = 1$.

Figure 2 shows the diffraction patterns of RS and BS sequences in the first Brillouin zone ($0 < q < 2\pi$). Inspection of these patterns shows that, although their average value is the same, the speckles repartition is remarkably different. In particular, it is clear that the BS pattern exhibits much more spikes, while the RS pattern is more homogenous and regular (no low-intensity speckles more

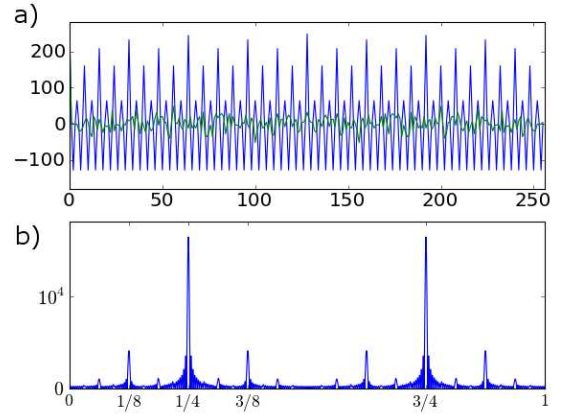


FIG. 4. a) Computed $g_4(n)$ using periodic boundary conditions for $N = 256$ RS sequence (blue) and a BS sequence (green). b) Magnitude of RS $\hat{g}_4(q)$ as a function of $h = q/2\pi$.

regularly spaced). To quantify this observation, we studied the statistics of both speckle patterns by calculating their probability density of intensity $P(I)$. It is known that for a random media (see *e.g.* [25]), the intensity distribution has a negative exponential distribution given by:

$$P(I) = \frac{1}{I} \exp\left(-\frac{I}{I}\right), \quad (9)$$

which in our case reduces to $P(I) = \exp(-I)$.

Figure 3a) shows the probability densities $P(I)_{RS}$ and $P(I)_{BS}$ for the RS and BS sequences. While $P(I)_{BS}$ follows quite well the negative exponential law, as expected, it is not the case for $P(I)_{RS}$. Though the precision of $P(I)_{RS}$ depends on N , it is well approximated by the step function $P(I < 2) = 0.5$. This statistics, which means that intensities lower than 2 occur with the same probability, explains the homogeneous aspect of the diffraction pattern. The reason for this unusual statistics is not clear, but undoubtedly comes from the GO nature of the RS sequence. The presence of order, invisible through diffuse scattering, is revealed by the statistics of the speckle pattern, breaking the D-homometry in a quantifiable way.

In order to test the robustness of the $P_{RS}(I)$ behavior with respect to disorder, we first quantify the degree of order of the RS by one of its quadruplet CF:

$$g_4(n) = \overline{\sigma_0 \sigma_1 \sigma_n \sigma_{n+1}}. \quad (10)$$

Indeed, we found numerically that at variance with the BS, $g_4(n)$ is LRO for the RS sequence (Fig. 4a)[26]. This is confirmed by the behavior of its FFT $\hat{g}_4(q)$ (Figure 4b), which exhibits well defined peaks indexed by the basis vectors $\{\frac{h_i}{4.2}, |h_i| \in \{-1, 1\}, i \in N\}$ [27] characteristic of limit-periodic functions [28]. By analogy with two-point orders, we define $\eta_4 \equiv \hat{g}_4(\pi/2)/N^2 = 1/4$ as the order parameter of this sequence.

We first decreased η_4 while keeping g_4 -LRO and g_2 -disorder by using the "Bernoullization" procedure as defined in [12]. It consists in changing the sign of each σ_n with probability p , in order to build sequences intermediate between the pure RS ($p = 0, 1$) and BS ($p = 0.5$) sequences. The order parameter η_4 was found to vary as $\eta_4(p) \simeq (1 - 2p)^4/4$.

Typical density probabilities density shown in Fig. 3b), exhibit a continuous evolution as a function of p . The step-function behavior is rapidly lost as $p \rightarrow 0.5$, with the best sensitivity close to the small intensity values $P(I = 0) \equiv P_0$. Simulations show that $P_0(p)$ closely follows $1 - 2\eta_4(p)$, and that sizeable deviation from the normal law starts from $p \gtrsim 0.8$.

Effect of g_4 -SRO on the speckle pattern was studied by shifting the sequence by two lattice periods at certain points, randomly selected with probability p . This ensures to achieve g_4 -SRO, clearly seen by the broadening of the $\hat{g}_4(\pi/2)$ peak, while keeping the $g_2(n)$ correlation to zero. Figure 3c) shows the results for different p values, corresponding to the average distances between faults ξ given in the caption. We checked that $P(I)_{RS}$ is unaffected for $\xi \lesssim N/40$. This shows that the extent of the four-point order has also an effect on the density probability, much larger than in the Bernoullization procedure.

Finally, let us mention that the problem of C-homometry, which is somehow the *true* homometry, is clearly related to the unicity of inverse problems, which is beyond the scope of this paper. It is important to note, however, that the use of ptychography [5], in which diffraction patterns are obtained by shifting illumination on the sample, can solve difficult problems of phase retrieval [29], including C-homometry.

DISCUSSION

It might seem pointless to discuss the problem of homometry while phase retrieval algorithms and ptychography can provide the full structural information, including high-order correlation functions. However, the full measurement of 3D speckle patterns is time consuming and in many situations it is not possible to get the data needed for such inversions. Methods of speckle analyses on quickly measured diagrams are thus needed to get novel information on the materials.

As stressed in ref. [30], real cases of true B-homometry are rare. On the contrary, because disorder is concerned, D-homometry is very frequent especially when systems are large. Moreover, it is related to high-order CF, which are hardly accessible to experiments [8, 10, 31][32]. However, although D-homometric systems have almost always different speckle patterns, it is not yet clear whether this difference is quantifiable. Indeed, though the probability density $P_{RS}(I)$ of our test bed sequence is strikingly different from its homometric BS, the observed effects are

very sensitive to disorder, and are almost invisible when four-point correlation lengths are too short. In this respect, we checked that the probability density of the lattice shown in Fig. 1 does not present sizeable deviation from the normal law. Qualitatively, the "spikiness" of speckle patterns is quickly reinforced by the introduction of randomness, which makes deviation to the negative exponential curve delicate to observe.

Consequently, though it is tempting to conclude from this study that high-order correlation functions are directly observable through speckle statistics analysis, much theoretical work is still needed to find the *relevant* parameters controlling the statistics. Such an effort could be supported by more sophisticated analyses such as the use of second-order (or higher) probability functions [25]. The simple fact that the lattices of Fig. 1 can be reconstructed with minimum information shows that high-order CF are somehow hidden in the speckle repartition.

An experimental difficulty lies in the presence of two-point correlations in all real systems (the classical SRO), which could obviously mask the speckle distribution analysis. In this respect, we have checked that, at least for SRO lattices with no high-order correlations, dividing the speckles pattern intensity by its associated diffuse scattering one I_{DD} (by smoothing, averaging or fitting) makes $P(I/I_{DD})$ follow the normal decreasing exponential law. This can help disentangling high-orders effects from two-point ones.

Another issue might be the partial coherence of the beam, which reduces the speckles contrast and makes the previous analyses difficult. This could be overcome by the analysis of the speckles maximum intensities, which exhibit similar statistical properties (not shown here).

In conclusion, we suggest that speckle statistics analysis could be used as another tool to test for the presence of high-order correlations. Indeed much physics could be explored with the measurement of high-order correlations, still hidden to experiments (see ref. [31] for examples). We hope this work will impulse theoretical and experimental studies on the role of high-order correlation functions on coherent diffraction diagram.

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